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REFLECTED SHOCK INITIATION OF A CHEMICAL REACTION

by

Ralph F. Shear

MAY 1966

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BALLISTIC RESEARCH LABORATORIES

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REFLECTED SHOCK INITIATION OF A CHEMICAL REACTION

Ralph E. Shear

Computing Laboratory

RDT & E Project No. 1P014501A14B

ABERDEEN PROVING GROUND, MARYLAND

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MEMORANDUM REPORT NO. 1743

REShear/bj
Aberdeen Proving Ground, Md.
May 1966

REFLECTED SHOCK INITIATION OF A CHEMICAL PEACTION

ABSTRACT

The Lax finite difference method is used to compute the hydrodynamic flow which results from the initiation of a chemical reaction by a reflected shock wave. The chemical reaction is assumed to be irreversible and of first order; initial conditions are chosen such that negligible reaction occurs behind the incident shock front.

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LIST OF SYMBOLS

a^*	$= c^*/V^*$, Lagrangian sound speed
a	$= a^*/a_o^* = \dfrac{a^* V_o^*}{c_o^*}$
c^*	sound speed
c	$= c^*/c_o^*$
E^*	specific total energy (internal plus kinetic)
E	$= E^*/(c_o^*)^2$
L_o^*	a characteristic length
m	mass coordinate
p^*	pressure
p	$= p^*/p_o^*$
T^*	temperature
T	$= T^*/T_o^*$
t^*	time
t	$= c_o^* \ t^*/L_o^*$
u^*	flow velocity
u	$= u^*/c_o^*$
V^*	specific volume
V	$= V^*/V_o$
x^*	distance
X	$= x^*/L_o^*$

LIST OF SYMBOLS (Cont'd)

γ_o	specific heat ratio of species A
γ_1	specific heat ratio of species B
η	mass fraction of A

Superscript

*	denotes dimensional quantity

Subscript

o	denotes initial conditions in the unshocked species A.

INTRODUCTION

The study of chemical kinetics by reflected shock techniques has been the subject of many recent investigations[1-5*]. The major advantages of the reflected shock technique are that higher temperatures are attainable than with incident shock techniques and, secondly, the gas behind a normally reflected wave is nearly stationary, thus the reaction is more readily observed.

In kinetic studies, only error in the temperature is liable to cause serious errors in the determination of the reaction rates; thus, it is necessary to be able to infer temperature precisely from measured shock parameters. Numerous investigators have inferred shock temperatures behind reflected waves from measurements of pressure, density, etc., and have found that temperatures may be lower than theoretical by about 2 percent[5-7]. Strehlow and Case[3] and Rudinger[8], however, have found that temperatures behind the reflected wave may be slightly higher than the values computed from ideal, steady-state shock tube theory. Johnson and Britton[5] have demonstrated the existence of lower reaction rates -- which imply lower temperatures -- behind the reflected shock wave, whereas, Fishburne et al[9] have shown the existence of slightly higher reaction rates behind the reflected wave.

Strehlow and Cohen[4] have demonstrated the usefulness of reflected shock techniques in the study of initiation of detonations. They have observed the reaction wave behind the reflected shock and have noted that under certain conditions the reaction wave either developed into a detonation wave before interacting with the reflected wave, or overtook the reflected wave first and then developed into a detonation wave, or, finally, the reaction merely accelerated the reflected wave.

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*Superscript numbers denote references which may be found on page 31.

EQUATIONS

Assume that we have a semi-infinite tube extending from $x^* = -\infty$ to $x = L_o^*$ and that the tube contains a gaseous species A which is capable of undergoing the irreversible, exothermic reaction $A \rightarrow B$. Furthermore, assume that the initial pressure distribution to be the piecewise uniform state, p_o^* for $x^* > 0$ and $p_1^* > p_o^*$ for $x^* \leq 0$. The flow velocity u_o^* for $x^* > 0$ is assumed to be zero, the temperature to be T_o and specific volume to be V_o^*. Initial conditions for $x^* \leq 0$ are given by the ideal gas Hugoniot conditions. In our sample calculations we shall assume that $p_1^*/p_o^* = 4.5$; thus the incident shock speed, in units of the ambient sound speed c_o^* of species A at p_o^*, T_o^*, and of specific heat ratio, γ_o, of 7/5, is 2. The parameters in the assumed reaction rate equation are chosen such that negligible reaction occurs behind the incident shock front. Under these conditions, the incident wave will reach the closed end of the tube, $x^* = L_o^*$, at about $L_o^*/(2c_o^*)$ time units. The shock reflection results in an increase of temperature which causes further reaction to occur; this increased reaction results in the formation of a compressive wave which eventually overtakes the reflected wave. This process will be evident in the numerical solution of the partial differential equations which describe the reactive flow. See, for example, Figure 12.

The equations describing the non-steady, non-viscous, one-dimensional flow of a reacting fluid in which the irreversible, first order chemical reaction $A \rightarrow B$ takes place may be written in the Lagrangian form:

$$\frac{\partial V}{\partial t} = \frac{\partial u}{\partial m} \tag{1}$$

$$\frac{\partial u}{\partial t} = -\frac{1}{\gamma_o}\frac{\partial p}{\partial m} \tag{2}$$

$$\frac{\partial E}{\partial t} = -\frac{1}{\gamma_o}\frac{\partial (pu)}{\partial m} \tag{3}$$

$$\frac{\partial \eta}{\partial t} = -\nu\eta \exp\{-E^{\#}/T\} \tag{4}$$

$$\frac{\partial X}{\partial t} = u \tag{5}$$

where η is the mass fraction of species A, ν is a frequency factor, and $E^{\#}$ is the activation energy, in units of $(c_o^*)^2$. The remaining symbols have been defined previously. (page 7).

In addition to these equations, we have the equations of state

$$p = p(I,V,\eta), \tag{6}$$

where $I = E - \frac{1}{2}u^2$ is the specific internal energy, and

$$T = T(p,V). \tag{7}$$

In the following, we assume that both species A and B are polytropic gases with their respective internal energy functions given by

$$I_A = \frac{T}{\gamma_o(\gamma_o-1)} + I_A^{\#} \quad \text{and}$$

$$I_B = \frac{T}{\gamma_o(\gamma_1-1)} + I_B^{\#},$$

where γ_o, γ_1, $I_A^{\#}$ and $I_B^{\#}$ are constants. Furthermore, we assume that at any given value of p, V, and T the mixture of A and B obeys the ideal gas law

$$pV = T \tag{8}$$

and that the internal energy of the mixture of A and B is given by

$$I = \eta I_A + (1-\eta) I_B .$$

With these assumptions, equation (6) becomes

$$p = \left\{I - \left[\eta I_A^{\#} + (1-\eta)I_B^{\#}\right]\right\} \left[\frac{\gamma_o(\gamma_o-1)(\gamma_1-1)}{(\gamma_1-1)\eta + (1-\eta)(\gamma_o-1)}\right] \left(\frac{1}{V}\right) \tag{9}$$

and T is given by equation 8.

DIFFERENCE EQUATIONS

The partial differential equations (1) - (5) must be replaced by a system of difference equations in order to obtain a numerical solution. The equations (1) - (5) are all of the form

$$\frac{\partial y}{\partial t} + \frac{\partial F}{\partial m} + B = 0$$

where F and B are functions of m, t and y. Such equations are called conservation laws and are of the form used in the difference method proposed by Lax[10] in 1954. In the Lax method time derivatives y_t are replaced by

$$\frac{\partial y}{\partial t} \approx \frac{y_n^j - \frac{1}{2}\left(y_{n-1}^{j-1} + y_{n+1}^{j-1}\right)}{\Delta t}$$

and space derivatives F_m by

$$\frac{\partial F}{\partial m} \approx \frac{F_{n+1}^{j-1} - F_{n-1}^{j-1}}{2\Delta m}$$

where superscript j denotes the value of the variable at time, $t = t^{j-1} + \Delta t^j$ and subscript n refers to the spatial position $m = n\Delta m$. The spatial grid size Δm is fixed whereas the time-step depends upon j. Letting $\overline{F} = \frac{1}{2}\left(F_{n+1}^{j-1} + F_{n-1}^{j-1}\right)$ the difference equations approximating equations (1)-(5) become

$$V_n^j = \overline{V} + \frac{\lambda^j}{2}\left(u_{n+1}^{j-1} - u_{n-1}^{j-1}\right) \tag{10}$$

$$u_n^j = \overline{u} - \frac{\lambda^j}{2\gamma_o}\left(p_{n+1}^{j-1} - p_{n-1}^{j-1}\right) \tag{11}$$

$$E_n^j = \overline{E} - \frac{\lambda^j}{2\gamma_o}\left[(pu)_{n+1}^{j-1} - (pu)_{n-1}^{j-1}\right] \tag{12}$$

$$\eta_n^j = \overline{\eta} - \nu\Delta t^j\,\overline{\eta}\exp\{E^{\#}/\overline{pv}\} \tag{13}$$

$$X_n^j = \bar{X} + \frac{\Delta t^j}{2}\left[u_n^j + \bar{u}\right] \tag{14}$$

$$I_n^j = E_n^j - \frac{1}{2}\left(u_n^j\right)^2 \tag{15}$$

$$p_n^j = \frac{1}{V_n^j}\left[\frac{\gamma_o(\gamma_o-1)(\gamma_1-1)}{(\gamma_1-\gamma_o)\,\eta_n^j + (\gamma_o-1)}\right]\left[I_n^j - \left\{\eta_n^j I_A^{\#} + (1-\eta_n^j)I_B^{\#}\right\}\right] \tag{16}$$

where $$\lambda^j = \Delta t^j/\Delta m.$$

The ratio $\lambda^j = \Delta t^j/\Delta m$ is calculated from the stability condition that

$$\lambda^j = \frac{b}{\max\limits_n \left\{a_n^{j-1}\right\}} \qquad \text{for } 0 < b \le 1$$

and where a, the Lagrangian sound speed, is given by

$$a_n^j = \left[\frac{\gamma_1(\gamma_o-1) + (\gamma_1-\gamma_o)\eta_n^j}{\gamma_o\left\{(\gamma_1-\gamma_o)\eta_n^j + \gamma_o-1\right\}} \quad \frac{p_n^j}{V_n^j}\right]^{1/2}$$

INITIAL AND BOUNDARY CONDITIONS

In terms of the non-dimensional variables previously defined, the mass of gas A contained in the region $0 < X \le 1$ is equal to one. This region is divided into N equal mass zones, i.e.,

$$\Delta m = \frac{1}{N}\,;$$

hence, for this section the Lagrangian mass coordinate has, initially, the same numerical value as the Eulerian coordinate. The section $-\infty < X \le 0$ is replaced by a finite section containing M mass zones. The initial conditions are:

$$p_n^o = \begin{cases} p_1 & n \le 0 \\ 1 & 0 < n \le N \end{cases} \qquad a_n^o = (p_n^0/V_n^o)^{1/2}$$

$$V_n^o = \frac{p_n^o + 6}{6p_n^o + 1} \qquad X_n^o = \frac{n}{N} V_n^o$$

$$u_n^o = \frac{5(p_n^o - 1)}{\sqrt{7(6p_n^o + 1)}} \qquad F_n^o = \frac{p_n^o V_n^o}{\gamma_o(\gamma_o - 1)} + \frac{1}{2}(u_n^o)^2$$

$$\eta_n^o = 1. \qquad \gamma_o = 7/5$$

In the numerical example given here $p_1 = 4.5$, thus the shock speed, in units of the ambient sound speed of A, is 2.

The boundary condition that the particle velocity u be zero at the reflecting wall is readily incorporated into the difference method. This is accomplished by defining values at an imaginary mesh-point, (j, N+1), as follows: Let

$$u_{N+1}^j = - u_{N-1}^j$$

and all other values at (j,N+1) be equal to their respective values at (j,N-1); thus equation (11), at n=N, automatically gives $u_N^j = 0$. The remaining values to be specified are:

$$b = 1 \qquad E^{\#} = 25$$

$$\gamma_o = \gamma_1 = 7/5 \qquad I_A^{\#} = 0$$

$$\nu = 10^6 \qquad I_B^{\#} = - 26$$

RESULTS

The results of the numerical computation of equations (10) - (15) are summarized in Figures 1-7. Figure 1 is a space-time plot of selected particle paths and shows the general features of the flow. In particular, the incident and reflected shocks and the reaction wave are clearly evident. After interaction of the reaction wave and reflected wave occurs, a double wave appears. (The double wave is more evident in Figure 12). Each of the two waves appears to be traveling at nearly constant velocity. The secondary wave, receding from the first, is a shock wave traveling in gas B. The first wave appears to have all the characteristics of a detonation as the values of the pressure, etc, correspond to those expected from a detonation transition of species A into species B, under the chosen initial conditions. The observed double wave structure is similar to that observed by Cher and Kistiakowsky[11] in their photographic studies of detonation of certain hydrocarbon-oxygen mixtures. Cher and Kistiakowsky concluded that "the secondary wave is due to entropy increase in the rarefaction wave caused by a spontaneous reaction". Analysis of the wave patterns observed in these computations will be reported in subsequent reports, and it is expected that further details concerning the double wave structure will be reported at that time.

A typical pressure-distance curve, corresponding to a fixed time, is shown in Figure 2. This pressure profile is typical of the pressure wave after the occurrence of interaction between the reflected and reacting wave. Negligible reaction has occurred ahead of the primary wave (corresponds to peak pressure) and the conditions in this region are, approximately, the same as behind the initial incident shock front, i.e., $p = 4.5$, $u = 1.25$, etc. An ideal Chapman-Jouguet detonation wave traveling into species A at these conditions, and instantaneously converting A into B gives

$$p_{CJ} = 84.70 \qquad V_{CJ} = 0.2237$$

$$u_{CJ} = -1.694 \qquad D = -6.047 ,$$

where D denotes the speed of the wave. The computed values, corresponding to the peak pressure of Figure 2, are

$$p = 84.62 \qquad V = 0.2239$$

$$u = -1.692 \qquad D = -6.045$$

The tail of the pressure profile of Figure 2, i.e., the nearly constant pressure region adjacent to the wall, corresponds to the region where the particle velocity is approximately zero. If we assume that this region is connected to the detonation state by means of a rarefaction wave, the pressure ratio at the wall would be 48.02 as compared with the computed pressure ratio of 48.17. The wall pressure at $j = 1049$, 300 time-steps later, is 48.02 and the quiescent region occupies a corresponding greater region of the tube. Comparison of these results with G. I. Taylor's similarity solution[12] will be reported in subsequent reports.

If we assume that the region bounded by the wall, the reflected shock and the reaction wave is constant and, in particular, assume the values of pressure and velocity are those given by the ideal gas shock equations[13], then for an incident shock pressure of 4.5, the pressure and particle velocity in the reflected zone are $p = 15$ and $u = 0$. The Chapman-Jouguet detonation wave values would be

$$p_{CJ} = 198.1 \qquad V_{CJ} = 0.1004$$

$$u_{CJ} = -2.944 \qquad D = -7.403 .$$

For comparison, Figure 3 shows the pressure, at constant time, just prior to the interaction of reflected and reaction waves. The pressure, corresponding to the peak, is 199.7 and the corresponding values of u and V are

$$u = -3.059,$$

$$V = 0.1003,$$

and the reaction is traveling at speed $D \approx -7.504$.

Conditions behind the reflected wave, prior to the observed development of the reaction, are not constant as can be seen in Figures 4 and 5 which are, respectively, plots of the wall pressure and wall temperature versus time.

The reaction is complete, i.e., $\eta = 0$, at the time corresponding to the maximum pressure of Figure 3. The mass fraction, η, and the specific volume ratio, at $X = 1$, are shown, respectively, in Figures 6 and 7.

If ν in equation 12 is set equal to zero and the remaining parameters and conditions are unchanged, the problem considered is merely that of normal shock reflection in an ideal gas. For purposes of comparison and preliminary tests of accuracy ν was set equal to zero and the corresponding flow computed. In Figure 8 the particle paths are plotted. The results are those expected, i.e., the particle velocity is (nearly) zero behind the reflected wave and the pressure jump is that given by the well-known shock reflection formulas[13]. The pressure as a function of Eulerian distance is plotted for various grid sizes in Figure 9, and it is seen that the shock front becomes steeper as grid size decreases.

In Figures 10 and 11, percent relative errors in V and X are shown as functions of grid size. The errors in p and u are negligible, at these chosen points, for all grid sizes.

The effect of increasing the activation energy on the resulting reacting flow is illustrated in Fig. 12. The activation energy $E^{\#}$ was increased from 25 to 28.6. The values of the remaining parameters are the same as those used to obtain the results illustrated in Fig. 1.

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The author gratefully acknowledges the invaluable assistance received from Mr. Barry Rodin. Mr. Rodin was responsible for coding and programming this problem, complete with graphical procedures, on the BRLESC and the <u>Data Plotter</u>.

RALPH E. SHEAR

FIG. 1: REFLECTED SHOCK INITIATION OF AN EXOTHERMIC REACTION
COMPUTED PARTICLE PATHS
ACTIVATION ENERGY = 25

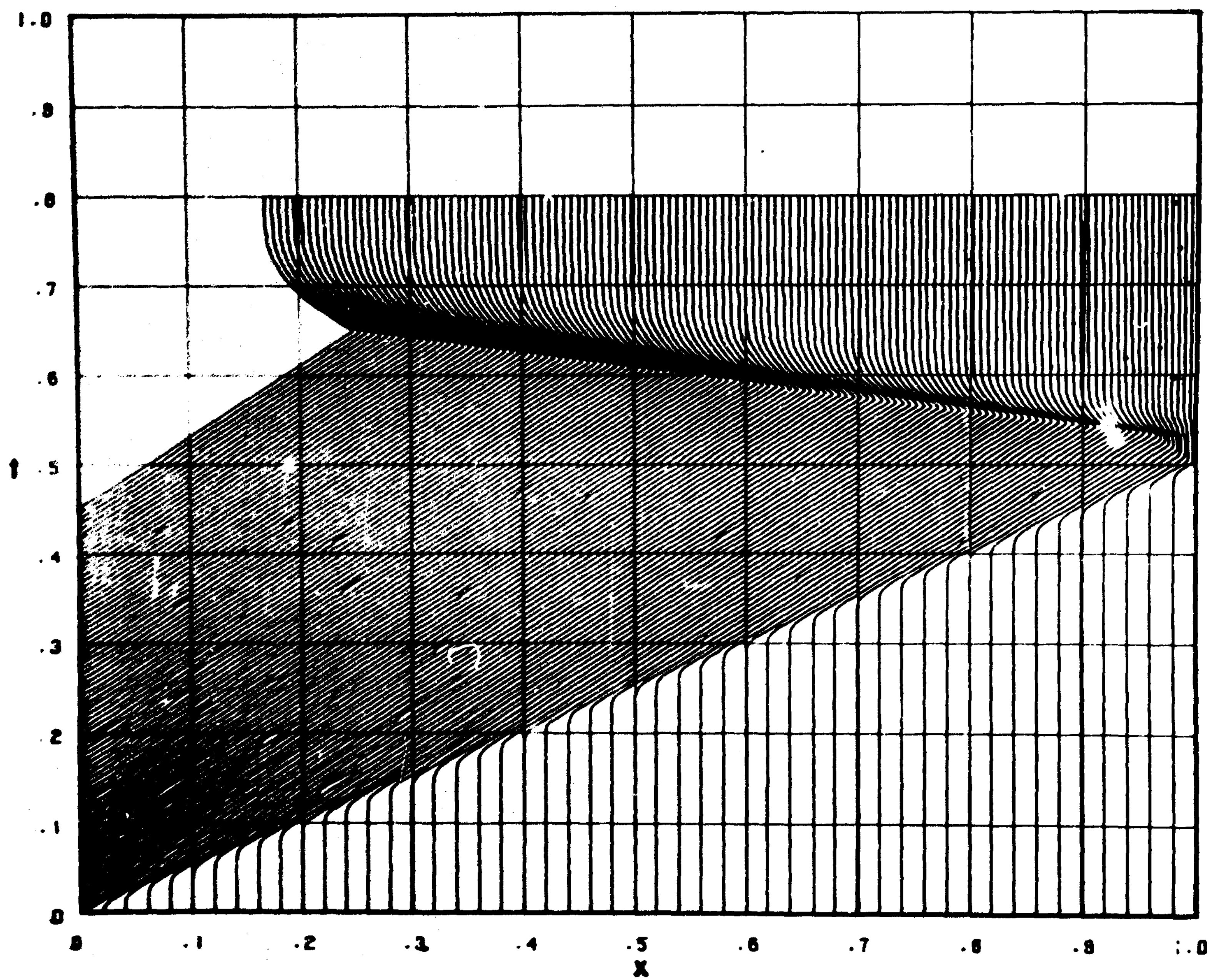


Fig. 2 Pressure Ratio vs Scaled Distance

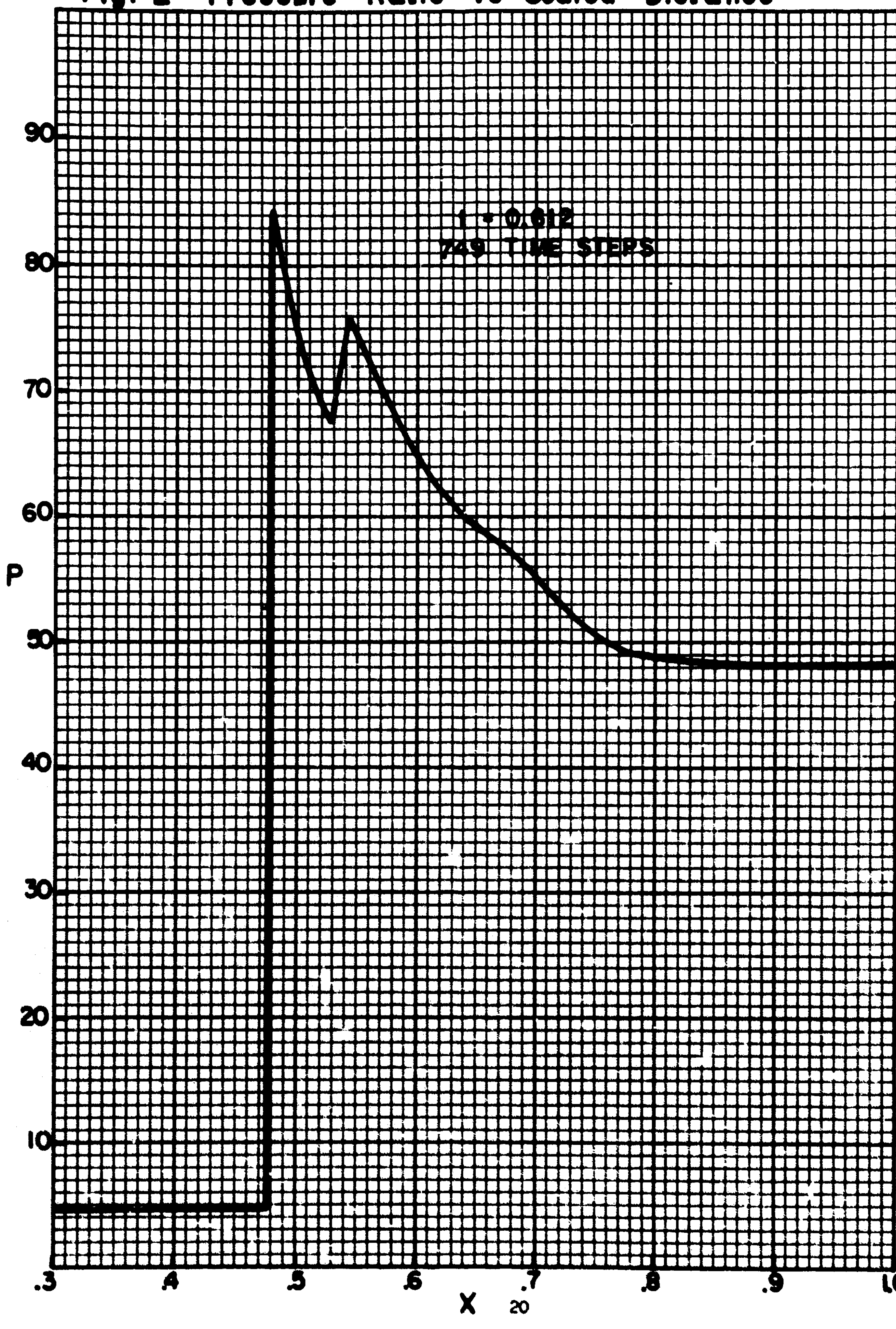


Fig. 3 Pressure Ratio vs Scaled Distance

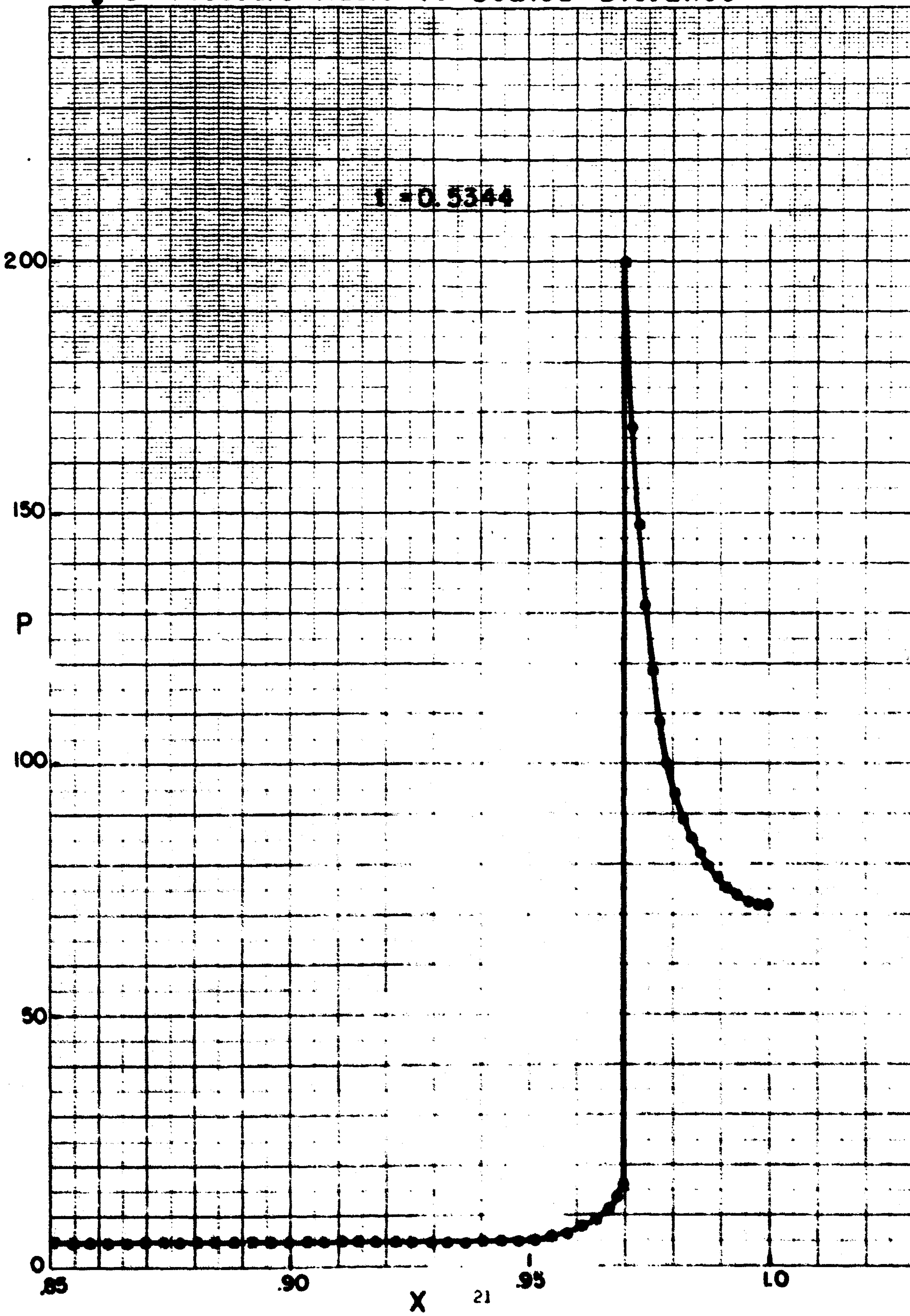


Fig. 4 Pressure at the Wall

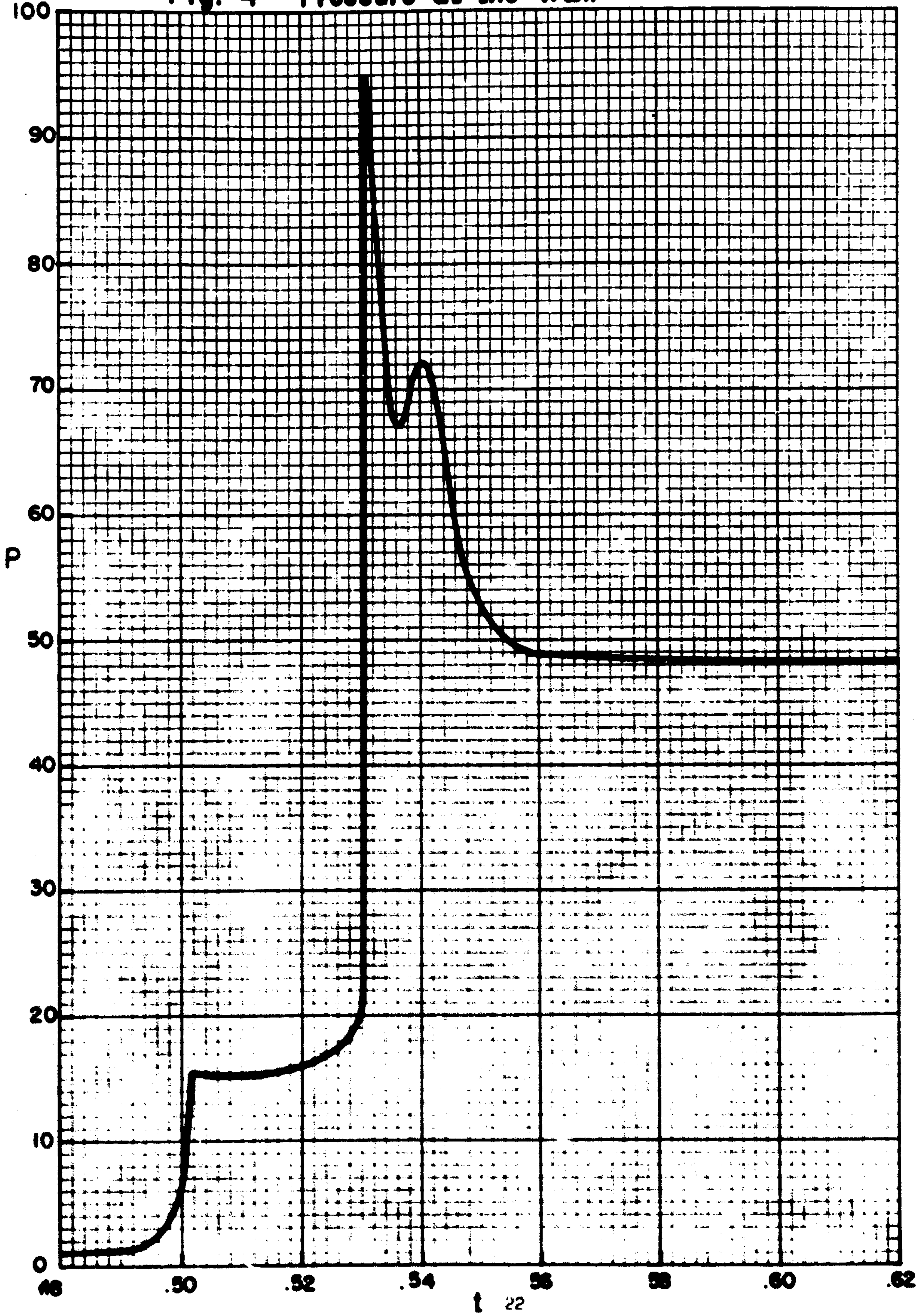


Fig. 5 Temperature at the Wall

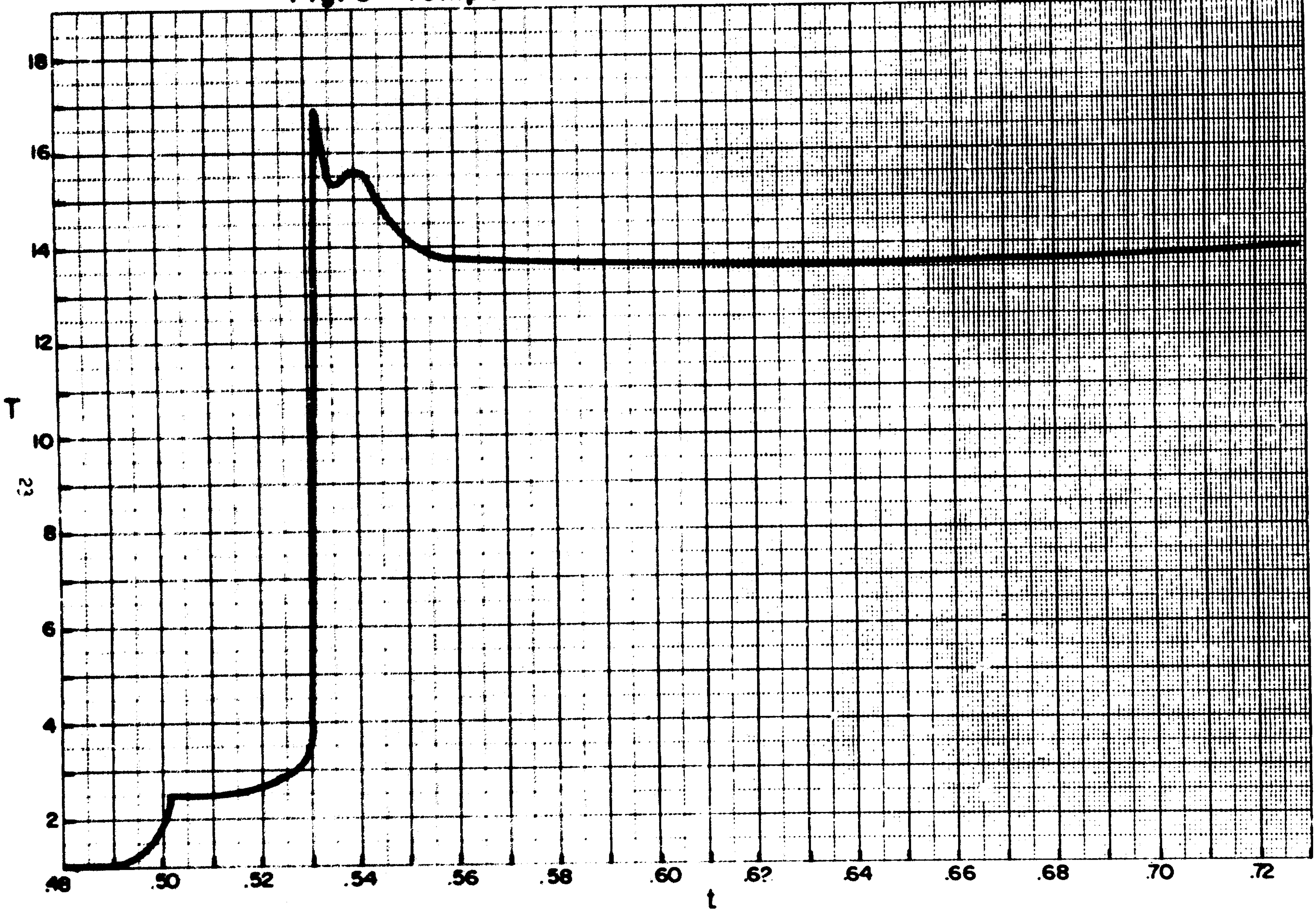


Fig. 6 : Mass Fraction of A at X = I vs Scaled Time

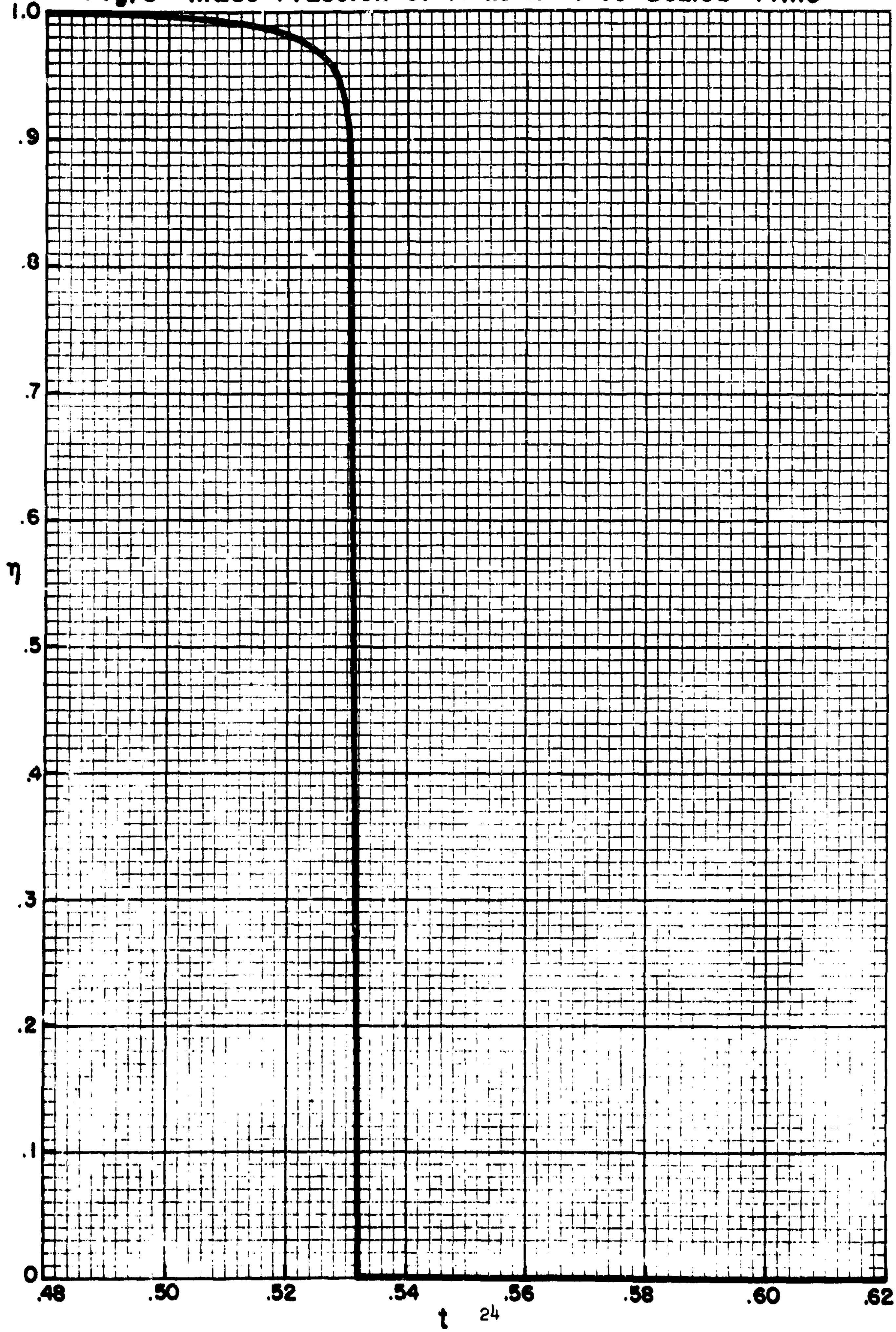


Fig.7 Specific Volume Ratio at X = 1 vs Time

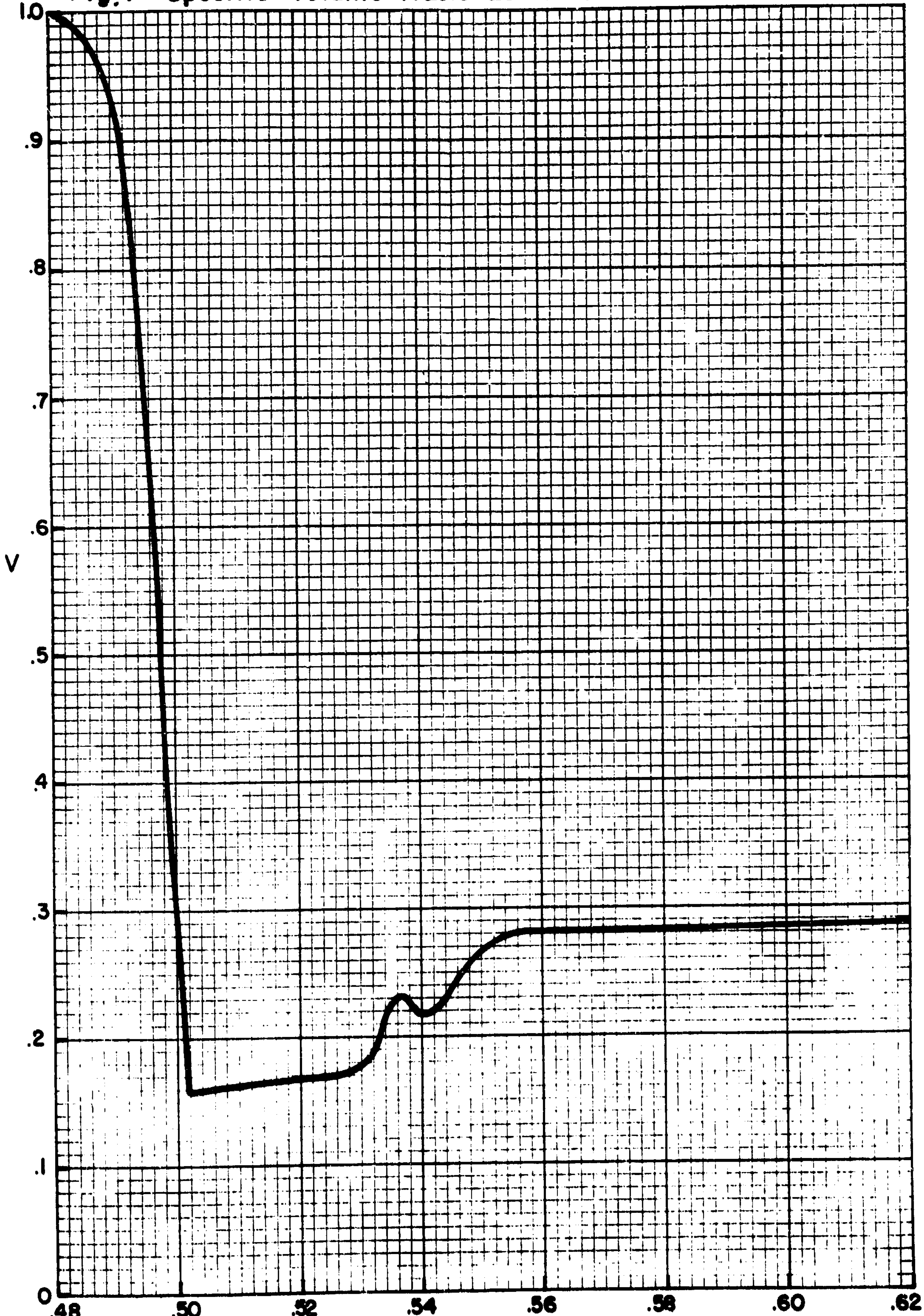


FIG. 8: REFLECTED SHOCK WAVE IN A NON-REACTING GAS
COMPUTED PARTICLE PATHS

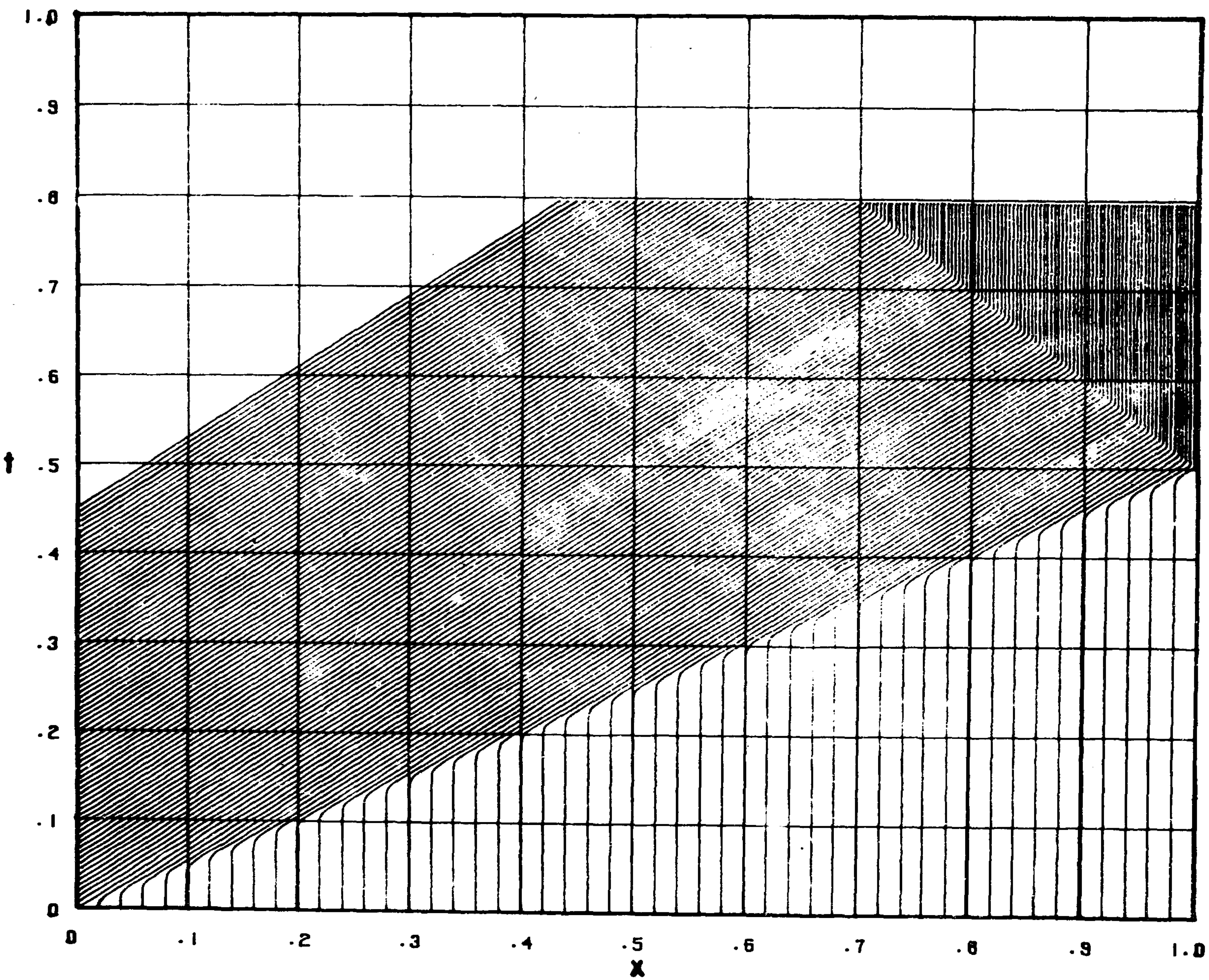


Fig. 9 Pressure vs Distance at t = 0.6 for non-reacting gas

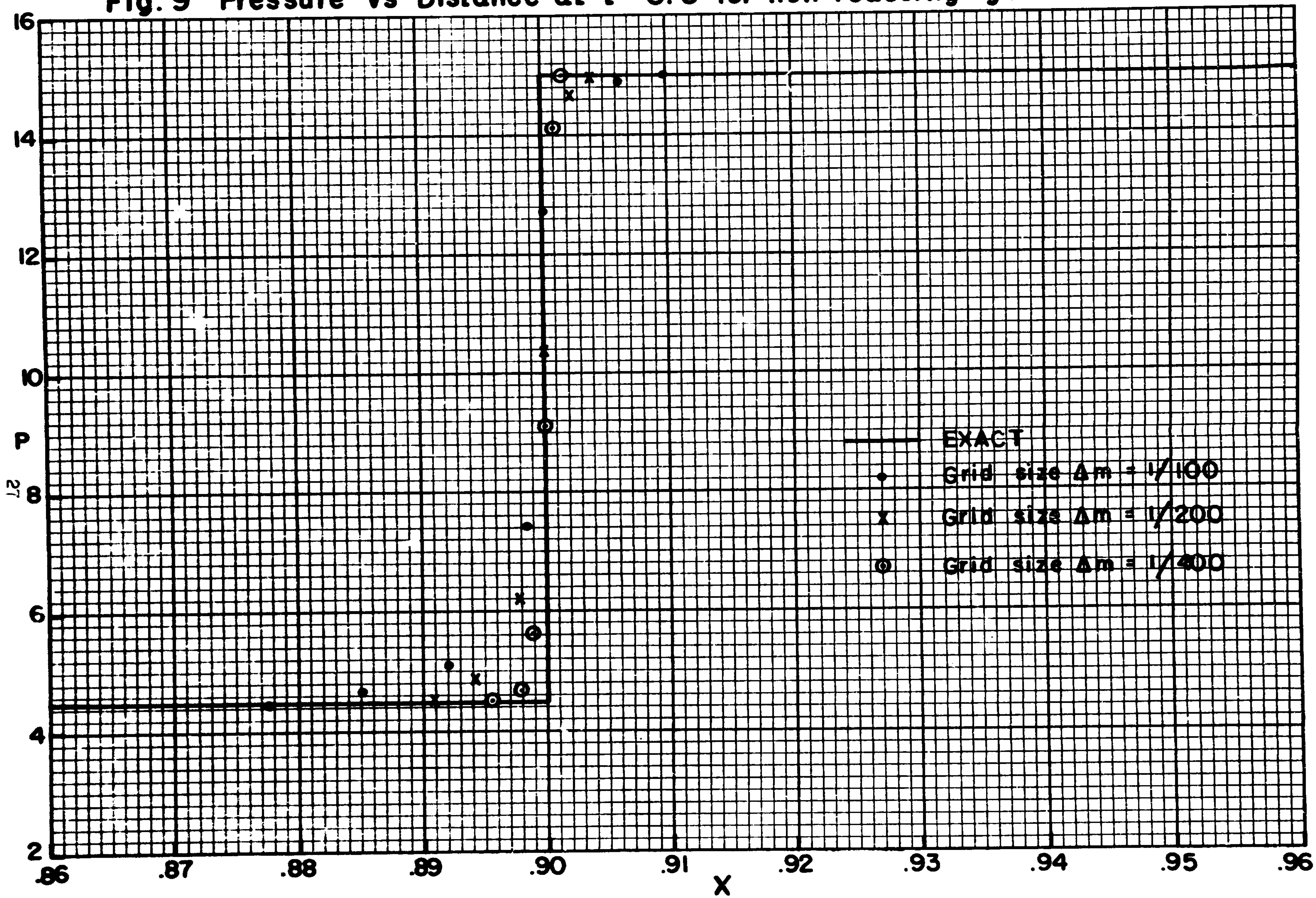


Fig. 10 Percent Relative Error in V and X at m = 0.15 and t = 0.6

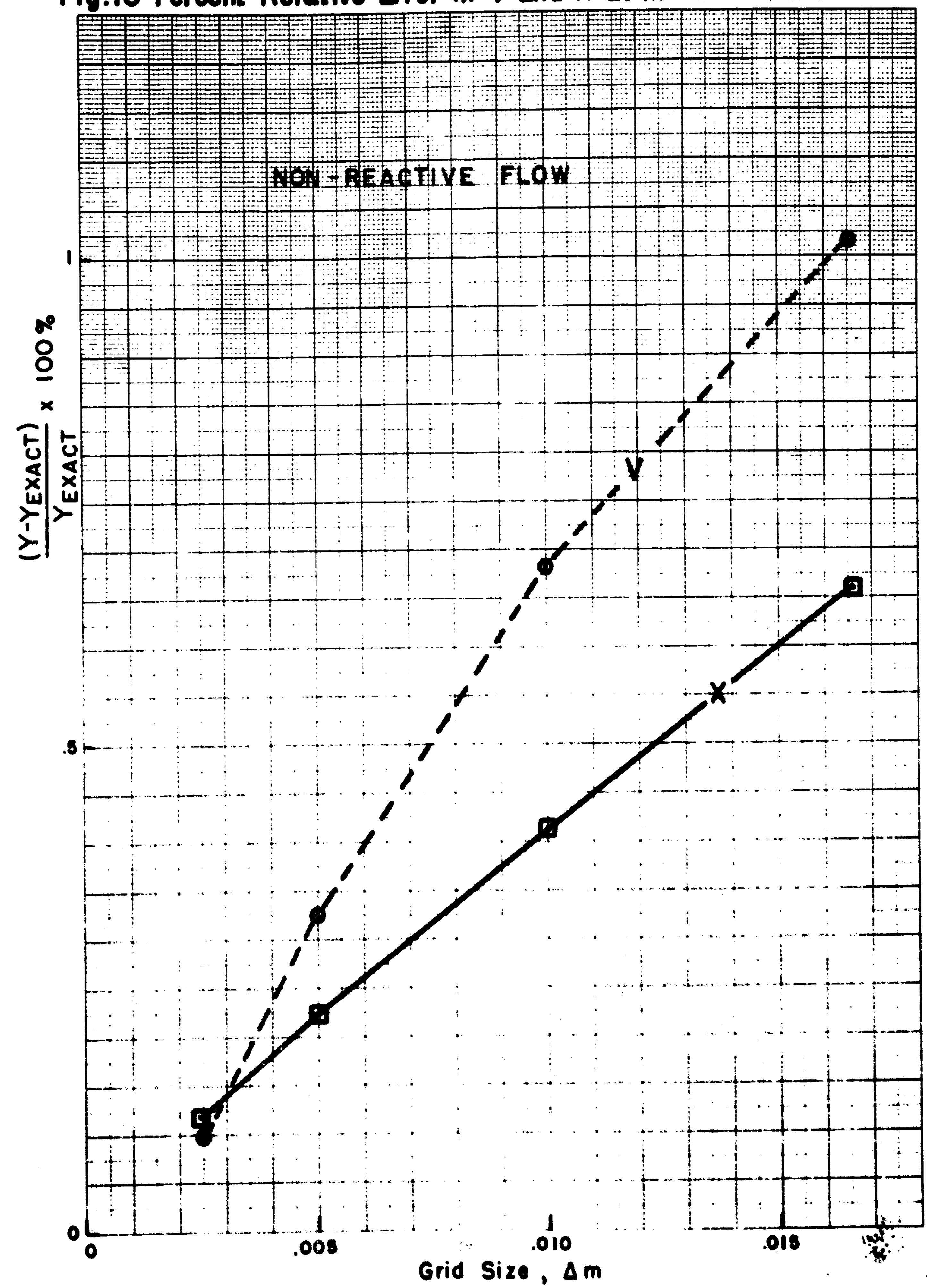


Fig. 11 Percent Relative Error in V and X at m = 0.75, t = 0.6

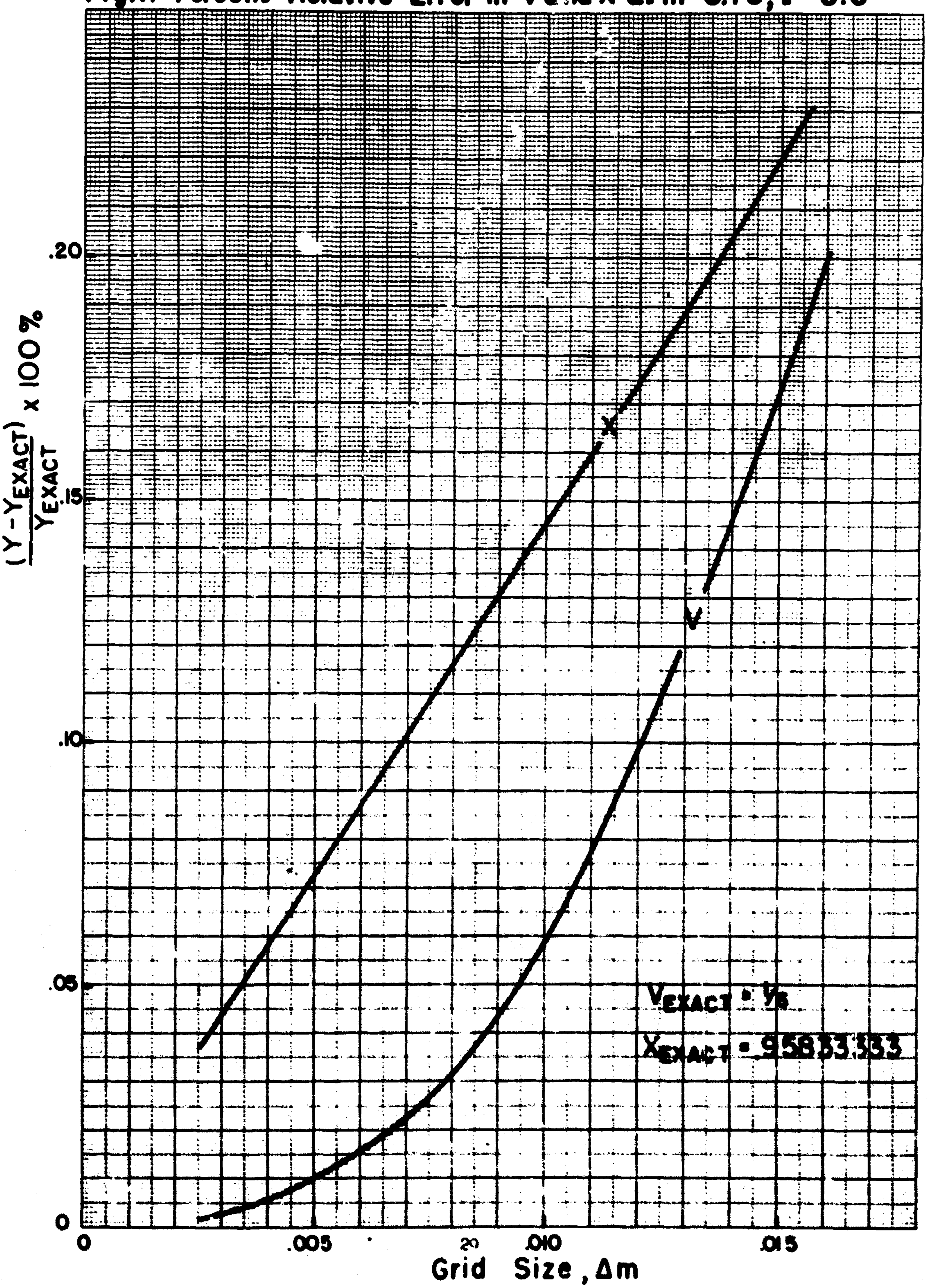
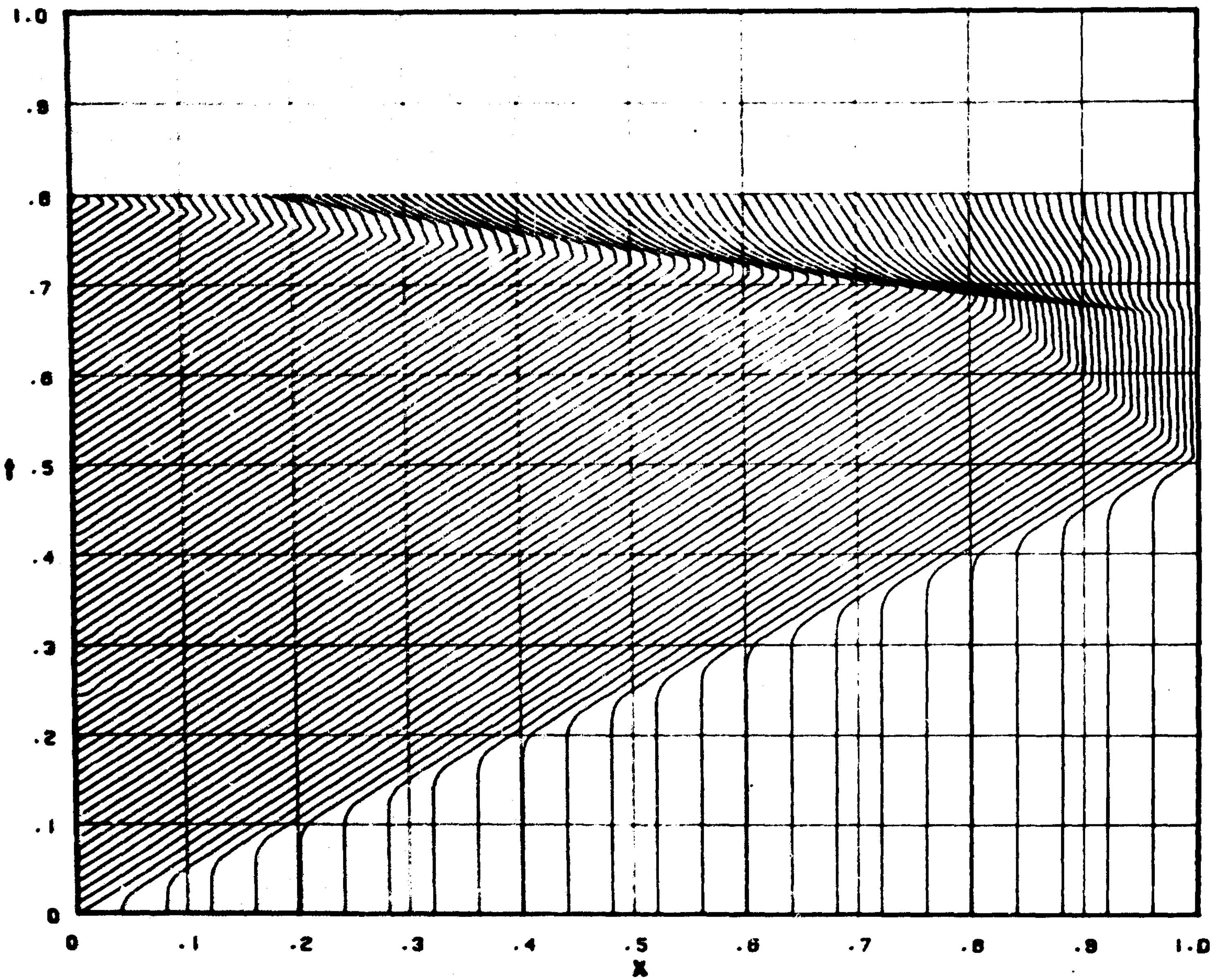


FIG 12: REFLECTED SHOCK INITIATION OF AN EXOTHERMIC REACTION
COMPUTED PARTICLE PATHS
ACTIVATION ENERGY = 28.6



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13. ABSTRACT

The Lax finite difference method is used to compute the hydrodynamic flow which results from the initiation of a chemical reaction by a reflected shock wave. The chemical reaction is assumed to be irreversible and of first order; initial conditions are chosen such that negligible reaction occurs behind the incident shock front.

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